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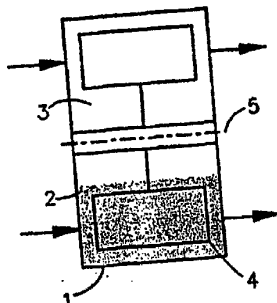
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(57) Abstract

The invention relates to a process for carrying out a reaction between a gas phase and a liquid phase in the presence of a monolithic catalyst, characterised in that the monolithic catalyst rotates around a horizontal shaft and is alternately in the gas phase and in the liquid phase. The invention also relates to an apparatus (1) in which a gas-liquid reaction can be carried out under the influence of a monolithic catalyst (4) rotating around a horizontal shaft (5).

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MONOLITHIC REACTOR

5           The invention relates to a process for carrying out a gas-liquid reaction in the presence of a monolithic catalyst.

          A process for carrying out a gas-liquid reaction in the presence of a monolithic catalyst is  
10   known from EP-A-614 869, which describes how products of peroxidic ozonolysis are reduced in a solution containing hydrogen in the presence of a monolithic catalyst. In one embodiment of the known process the monolithic catalyst is contained in the solution and  
15   hydrogen is passed through with stirring. In another embodiment of the known process the solution and the hydrogen are together contacted with the monolithic catalyst incorporated in a packed column. In a preferred embodiment of the process described in EP-A-  
20   614 869 the monolithic catalyst incorporated in the packed column is in a loop reactor.

          The drawback of the process described in EP-A-614 869 is that the gas-liquid mixture must be continuously circulated because, especially in the case  
25   of slow reactions, the residence time of the gas-liquid mixture in the column is too short to allow the reaction to go to completion.

          The aim of the invention is to provide a process that does not show this drawback.

30           This aim is achieved in that the monolithic catalyst rotates around a horizontal shaft and is alternately in the gas and in the liquid phase.

          This ensures that even slow reactions go to completion without it being necessary to continuously  
35   circulate the gas-liquid mixture.

          An advantage of such a process is that the good wetting of the monolithic catalyst enables better temperature control.

          A 'monolithic catalyst' is in this

specification understood to be a monolith bearing a catalyst. A 'monolith' is in this specification understood to be a one-piece catalyst carrier that is characterised by a structure with a large internal  
5 surface.

The monolith may in principle consist of any solid material. Examples are metal, metal oxide, ceramic, glass, plastic and carbon.

The carrier's large surface may consist of a  
10 large number of straight channels having a diameter between approx. 0.5 and 10 mm. The channels' cross section may vary considerably, depending on the type of material used. Known are e.g. round, square, hexagonal, triangular and T-shaped cross-sections and channels  
15 with fins on the inside.

The catalyst is applied to the channel walls by means of for example vapour deposition or impregnation, which results in a very large catalyst surface. In Catal.Rev.-Sci. Eng., 26(2), 179-270  
20 (1994), Cybulski and Moulijn describe how a monolithic catalyst can be prepared.

In the process according to the invention the monolithic catalyst revolves around a horizontal shaft and is alternately in the gas phase and in the liquid  
25 phase, so that gas and liquid alternately flow through the channels. This ensures that the gas need only diffuse through a very thin liquid film before a reaction with the liquid can take place at the catalyst surface. A good flow through the channels is obtained  
30 when the latter are in a plane perpendicular to the horizontal shaft. An even better flow through the channels can be obtained by fitting the ends of the monolithic catalyst facing away from the shaft with vanes so that they guide the liquid to the channel  
35 inlets.

An advantage of the process according to the invention is that the period of time for which the gas

diffuses through the liquid film can easily be varied via the choice of a rotation speed and a liquid level. Especially when the gas dissolves poorly in the liquid, lengthening this period of time may be advantageous to ensure that the gas-liquid reaction at the catalyst surface has proceeded to a sufficient extent before the liquid containing the reaction products is replaced. In this respect the monolithic catalysts incorporated in packed columns described in EP-A-614 869, which will hereinafter be referred to as "static monolithic catalysts", present the drawback that the ratio of the gas and the liquid that are contacted with the monolithic catalyst cannot be chosen freely: if there is too much gas, the liquid will not flow through some of the channels, while if there is too little gas the gas will not flow through some of the channels. With certain gas-liquid reaction rates the distribution of gas and liquid will be poor in the case of static monolithic catalysts owing to hydrodynamic instability; a large amount of gas will then flow through some channels while a large amount of liquid will flow through other channels. To obtain a desirable ratio it will often be necessary to circulate an undesirably large quantity.

Whenever reference is made to a 'gas-liquid reaction' in this specification this is understood to include not only a reaction between a gas and a liquid, but also between a gas and a substance dissolved in a liquid. This substance may be a gas, a liquid or a solid.

A monolith suitable for use as a monolithic catalyst may well have an internal surface of 2500 m<sup>2</sup>/m<sup>3</sup>. If so desired, monoliths can be enlarged by connecting several monoliths in the form of blocks, as a result of which the size of the desired monolith is not limited by the size of a producible monolith.

The type of catalyst to be used will depend

on the reaction to be carried out. In principle, all the known solid catalysts can be used as monolithic catalysts. Examples of suitable catalysts are the transition metals, their oxides, sulphides or mixtures  
5 hereof. Other examples are acid catalysts (e.g. aluminium or silicon oxide) or basic catalysts (e.g. magnesium oxide). Mixtures of the aforementioned catalysts can also be used.

EP-A-384 905 describes a process in which use  
10 is made of a gas-liquid reactor fitted with a vertical column packed with a monolithic catalyst. The gas and the liquid can be fed through the reactor in an upward or a downward direction. If the liquid flows downwards, at least a portion of the gas can flow countercurrently  
15 upwards. A drawback of such a configuration, however, is that backmixing may take place. 'Backmixing' is understood to be the phenomenon that some portions of liquid with higher concentrations of reaction products are mixed with fresh liquid elements rich in reactants.  
20 This often adversely affects the productivity and the selectivity. Another drawback of complete or partial countercurrent flow is that no reaction can take place in the channels that contain exclusively gas or liquid, as a result of which the catalyst's effectiveness  
25 factor and hence the productivity decrease.

The invention also relates to an apparatus for carrying out a reaction between a gas and a liquid, in the presence of a monolithic catalyst, with one or more monolithic catalysts being connected to a  
30 horizontal shaft.

Such an apparatus is suitable for carrying out slow gas-liquid reactions in particular, without it being necessary to continuously circulate a gas-liquid mixture.

35 Preferably the apparatus according to the invention is used in a configuration in which several apparatuses according to the invention are arranged in

these particles being damaged. Examples of such reactions are oxidation reactions under the influence of air.

Other characteristics and advantages will become apparent from the following description, in which reference is made to the appended drawings, in which:

- Figure 1 is a side view of the apparatus according to the invention, hereinafter referred to as "reactor", parallel to the monolithic catalyst's axis of rotation; Figure 2 is a side view of the reactor perpendicular to the monolithic catalyst's axis of rotation; Figure 3 is an enlarged cross-section of part of the monolith;
- Figure 4 is an enlarged longitudinal section of part of the monolith;
- Figure 5 is a side view of a series reactor parallel to the monolithic catalyst's axis of rotation.

In Figure 1, 1 is a reactor partly filled with liquid 2 and partly filled with gas 3. The reactor contains several monolithic catalysts 4 connected to a horizontal shaft 5. Figure 2 shows a round reactor 1 containing eight monolithic catalysts 4, three of which are to varying extents submerged in the liquid 2 while the other 5 are in the gas 3. Figure 3 shows part of the monolith, consisting of the carrier 6 with channels 7. The surface of the channels is covered with a catalyst, not visible in the drawing, on which there is a thin liquid film 8. Otherwise the channels are filled with the gas 3. Figure 4 shows a longitudinal section of the channels, whose surface is covered with the catalyst (not visible in the drawing) and a thin liquid film 8. In Figure 5 several reactors are in succession connected to a horizontal shaft.

C L A I M S

1. Process for carrying out a reaction between a gas phase and a liquid phase in the presence of a monolithic catalyst, characterised in that the monolithic catalyst rotates around a horizontal shaft and is alternately in the gas phase and in the liquid phase.  
5
2. Apparatus for carrying out a reaction between a gas and a liquid in the presence of a monolithic catalyst, characterised in that one or more monolithic catalysts are connected to a horizontal shaft.  
10
3. Apparatus according to Claim 2, characterised in that several apparatuses are arranged in series.  
15
4. Use of an apparatus according to Claim 3, with the gas and the liquid flowing countercurrently through the apparatus.
5. Use of the apparatus according to any one of Claims 2-4 in the catalytic hydrogenation or oxidation of a substance in a liquid phase.  
20
6. Use of the apparatus according to any one of Claims 2-4 in the execution of a bioreaction, in which the monolithic catalyst contains immobilised biological matter.  
25



series. An advantage of such an apparatus, which here and hereinafter is referred to as a "series reactor", is that backmixing has a much smaller effect. Backmixing may occur in the gas phase or in the liquid phase or in both phases. A further advantage of a series reactor is that if the gas and the liquid flow through the series reactor countercurrently, this can lead to a higher yield of the reaction.

10 A further advantage of a series reactor in which countercurrent flow of gas and liquid is used is that there is no risk of the liquid being entrained by the gas. This not only prevents backmixing, but neither can any problems occur in checking the liquid level as with the static monolithic catalyst described above.

15 A "horizontal shaft" is in this specification understood to be a horizontal or virtually horizontal shaft. The use of a virtually horizontal shaft in the series reactor can ensure that the gas-liquid ratio changes over the length of the series reactor. This may be advantageous in the case of reactions in which the amount of one of the two components present has at the end of the reaction decreased more than that of a different component. If the series reactor is placed not entirely horizontal, a lower concentration of for example the gas can be compensated because the volume of the gas in the reactor increases towards the end of the reaction.

The invention also relates to the use of the apparatus according to the invention in the catalytic hydrogenation or oxidation of a substance in a liquid phase. Characteristic of these reactions is that the hydrogen or oxygen to be used dissolves poorly in liquids, which is disadvantageous for the reaction rate, which under such conditions is limited by the diffusion of the gas through the liquid layer. To increase the concentration of gas in the liquid, and hence the reaction rate, these reactions are therefore

often carried out in the known reactors at elevated pressure. The greater amount of time taken by the liquid to flow from the channels of the catalyst monolith in the apparatus according to the invention  
5 leads to the formation of a much thinner liquid film on the catalyst surface than in the case of a static monolithic catalyst. The transport of gas particles is consequently less limited by diffusion, as a result of which the reaction rate is higher.

10 Usually a large amount of heat is released in hydrogenation and oxidation reactions. A drawback of the known apparatus for carrying out these reactions is that a local overconcentration of reactants is easily formed in a gas-liquid mixture, which may lead to an  
15 uncontrolled temperature increase. This may result in a hotspot or even a runaway. An advantage of the use of the apparatus according to the invention for a hydrogenation or oxidation reaction is that the entire catalyst surface is frequently submerged, which  
20 prevents the formation of dry, hot spots.

Other reactions for which the apparatus according to the process can be used with the same advantages as mentioned above are amination, halogenation and carbonylation.

25 The invention also relates to the use of the apparatus according to the invention in gas-liquid reactions in which biological matter like enzymes, yeasts or live cells is used as the catalyst. That the use of monoliths may present advantages in immobilising  
30 biological matter is known from EP-A-121,981, in which a feed liquid is circulated past tissue cells immobilised on a monolith, in which a chemical reaction takes place. The feed liquid may also contain solid particles. An advantage of the use of the apparatus  
35 according to the invention in biological reactions is that there is no need to circulate any solid particles in a solution, which means that there is less risk of

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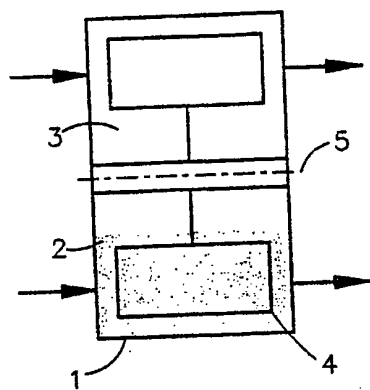


FIG. 1

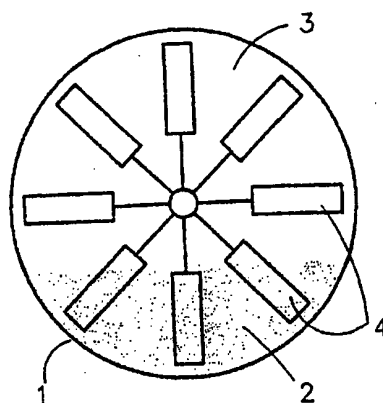


FIG. 2

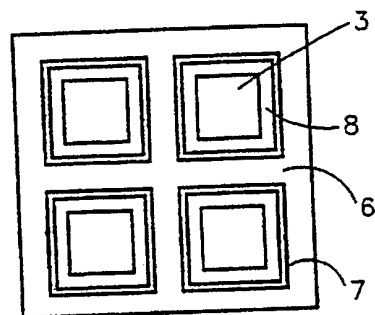


FIG. 3

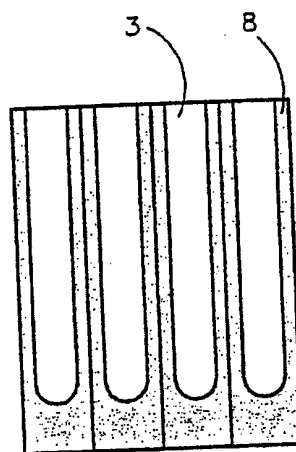


FIG. 4

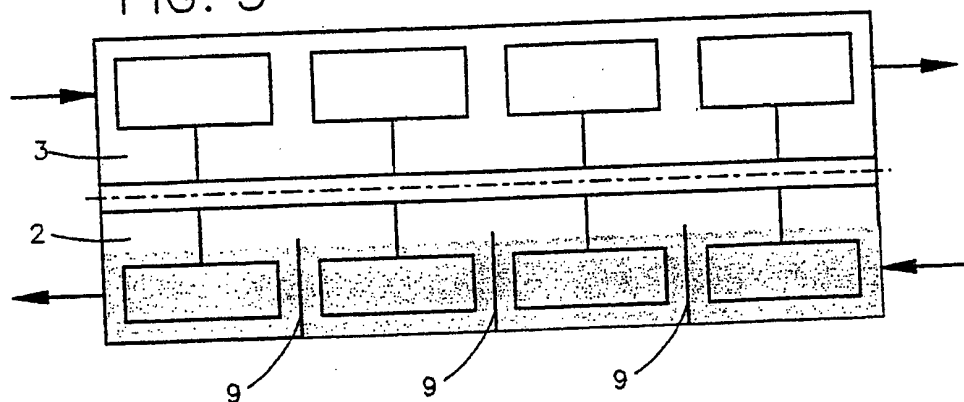


FIG. 5

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/NL 98/00011

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 B01J10/02 B01J10/00 B01J19/18

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

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## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 42 41 451 A (F.D.OESTE ET AL.) 16 December 1993 see abstract see page 3, line 35 - line 37 see page 7, line 58 - page 8, line 4 see figures 1,2	1,2
A	W0 91 09934 A (J.E.KITCHIN & S.P.DAVIES) 11 July 1991 see the whole document	1,2,6
A	GB 2 088 243 A (RTL CONTACTOR HOLDING S.A.) 9 June 1982	

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

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